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Vapor Pressure of Hydrofluoroolefins: Critical Review of Experimental Data and Models

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ABSTRACT

This paper critically reviews vapor pressure data and vapor pressure models for seventeen hydrofluoroolefins, including R1234yf and R1234ze(E), and presents Wagner-type vapor pressure correlations for the seventeen hydrofluoroolefins.

1. INTRODUCTION

In recent years there has been increasing commercial interest in low GWP refrigerants, particularly driven by the European Union's F-Gas Regulations and Mobile Directive. In particular, the Mobile Directive targets refrigerants with GWP > 150, including R134a. The current commercial focus is for R1234yf to replace R134a in automotive applications (*e.g.*, SAE 2009), primarily because of its low 100-year GWP relative to CO₂ of 4 (Nielsen *et al.*, 2007). There also is commercial interest in R1234yf for other refrigeration and air conditioning applications. In addition to R1234yf, one other fluorinated propene isomer, namely R1234ze(E), is receiving commercial interest as a refrigerant, blowing agent, and aerosol (EPA, 2012). It has a 100-year GWP relative to CO₂ of 6 (Søndergaard *et al.*, 2007). There also is considerable activity by chemical manufacturers and research groups to develop refrigerant blends with low GWP. Many of these blends are based on one or more of several fluorinated propene isomers (*e.g.*, R1234yf, R1234ze(E), R1243zf, to name a few) in combination with more traditional refrigerants such as R32, R125, R134a, R152a, and CO₂, among others. The purpose of these blends is to "tailor" the refrigerant to the particular application, while balancing factors such as GWP, flammability, toxicity, cost, thermophysical properties, COP, and volumetric capacity, to name a few.

2. REVIEW OF PUBLIC DOMAIN EXPERIMENTAL DATA

Since this paper focuses only on the vapor pressures of fluorinated propene isomers, the following review of publicly available experimental property data will limit its discussion to vapor pressure. Furthermore, since for the most part, R1234yf and R1234ze(E) are the two fluorinated propene isomers with thermophysical property data widely available in the public domain, the discussion will be limited to these two isomers.

For R1234yf, Di Nicola *et al.* (2010a), Fedele *et al.* (2011), Hulse *et al.* (2009), Richter *et al.* (2011), and Tanaka and Higashi (2010) have all presented vapor pressure data. In total, there are 125 data points over the saturation temperature range from 224 K to 363 K. As pointed out by Fedele *et al.* (2011), the 12 data points of Hulse *et al.* (2009) have mean absolute relative errors greater than 1 % when compared with all of the publicly available models

(discussed in the next section), excepting their own with which they have a mean absolute relative error of approximately 0.24 %. Therefore, the data of Hulse *et al.* (2009) should be used with caution, leaving 113 reliable publicly available data points.

For R1234ze(E), Di Nicola *et al.* (2012), Grebenkov *et al.* (2009), Kayukawa and Fujii (2009), McLinden *et al.* (2010), and Tanaka *et al.* (2010) have all presented vapor pressure data. In total, there are 168 data points over the saturation temperature range from 223 K to 380 K. As pointed out by Di Nicola *et al.* (2012), the 49 data points of Grebenkov *et al.* (2009) have mean absolute relative errors greater than 1.8 % when compared with all of the publicly available models, excepting their own with which they have a mean absolute relative error of approximately 1.2 %. Furthermore, Di Nicola *et al.* (2012) pointed out that the 32 data points of Kayukawa and Fujii (2009) have mean absolute relative errors greater than 0.72 % when compared with all of the publicly available models. Therefore, the data of Grebenkov *et al.* (2009) and Kayukawa and Fujii (2009) should be used with caution, leaving 87 reliable publicly available data points.

3. REVIEW OF PUBLIC DOMAIN VAPOR PRESSURE MODELS AND EQUATIONS OF STATE (EOS)

For R1234yf, Di Nicola *et al.* (2010a), Fedele *et al.* (2011), and Tanaka and Higashi (2010) provide Wagner-type vapor pressure correlations, and Fedele *et al.* (2011), Hulse *et al.* (2009), and Leck (2009) provide extended Antoine vapor pressure correlations. In addition to these, several EoS have been published, namely, Di Nicola *et al.* (2010b) and Leck (2009) present a Martin-Hou (M-H) EoS; Hulse *et al.* (2009) and Akasaka *et al.* (2010) present an Extended Corresponding States (ECS) EoS; Akasaka *et al.* (2010) present a Patel-Teja (P-T) EoS; Brown *et al.* (2009) and Brown *et al.* (2010) present a Peng-Robinson (P-R) EoS; and Akasaka (2011) and Lemmon *et al.* (2010) present a FEQ Helmholtz EoS.

For R1234ze(E), Di Nicola *et al.* (2012) and Tanaka *et al.* (2010) provide Wagner-type vapor pressure correlations, and Di Nicola *et al.* (2012) and Grebenkov *et al.* (2009) provide extended Antoine vapor pressure correlations. In addition to these, several EoS have been published, namely, Grebenkov *et al.* (2009) and Akasaka (2010) present an ECS EoS; Brown *et al.* (2010) present a P-R EoS; and Akasaka (2011), Lemmon *et al.* (2010), and McLinden *et al.* (2010) present a FEQ Helmholtz EoS.

In addition to the vapor pressure models and EoS for R1234yf and R1234ze(E), which are based on experimental data, Brown *et al.* (2010) present P-R EoS for eight fluorinated propene isomers, which include R1234yf and R1234ze(E). The additional six isomers are: R1225ye(E), R1225ye(Z), R1225zc, R1234ye(E), R1234ze(Z), and R1243zf. The P-R EoS for these isomers rely on known normal boiling point temperatures (NBP) and group contribution estimations for the critical state properties, ideal gas specific heats at constant pressure, and acentric factors.

4. VAPOR PRESSURE MODELS FOR 17 FLUORINATED PROPENE ISOMERS

In this paper, we provide Wagner-type vapor pressure correlations for seventeen fluorinated propene isomers (four isomers of R1225, seven isomers of R1234, and six isomers of R1243) listed in Table 1.

Table 2 list NBP for the isomers of Table 1. Amongst the 17 isomers, two (R1234yf and R1234ze(E)) are well-described with experimental data. Seven others have NBP reported in the public domain, which allows for good estimates of critical state properties, ideal gas specific heats at constant pressure, and acentric factors from which simple EoS can be constructed to yield quite reasonable estimates of thermodynamic properties (see, *e.g.*, Brown, 2007). For the other eight isomers of Table 1, estimates for the NBP have been made from which estimates of the critical state properties, ideal gas specific heats at constant pressure, and acentric factors are made and EoS are constructed.

In Table 2, the first column of data lists NBP from the primary public domain source (patents, patent applications, or referred journal articles). The second column of data lists NBP obtained from a leading chemical supplier, the third column of data lists NBP from NIST's webbook for two of the isomers, and the fourth column of data lists NBP from public domain sources, other than those previously listed, for three of the isomers.

Table 1: Seventeen fluorinated propene isomers for which some information is available in the public domain

Refrigerant	Molecular Formula	IUPAC Name	CAS Number	ChemSpider ID
R1225yc	$\text{CHF}_2\text{CF}=\text{CF}_2$	1,1,2,3,3-pentafluoroprop-1-ene	433-66-9	516398
R1225ye(E)	$\text{CF}_3\text{CF}=\text{CHF}$	(1E)-1,2,3,3,3-pentafluoroprop-1-ene	5595-10-8	4887279
R1225ye(Z)	$\text{CF}_3\text{CF}=\text{CHF}$	(1Z)-1,2,3,3,3-pentafluoroprop-1-ene	552843-8	4647379
R1225zc	$\text{CF}_3\text{CH}=\text{CF}_2$	1,1,3,3,3-pentafluoroprop-1-ene	690-27-7	62848
R1234yc	$\text{CH}_2\text{FCF}=\text{CF}_2$	1,1,2,3-tetrafluoroprop-1-ene	115781-23-2	10030466
R1234ye(E)	$\text{CHF}_2\text{CF}=\text{CHF}$	(1E)-1,2,3,3-tetrafluoroprop-1-ene	115781-19-6	10331119
R1234ye(Z)	$\text{CHF}_2\text{CF}=\text{CHF}$	(1Z)-1,2,3,3-tetrafluoroprop-1-ene	730993-62-1	10331120
R1234yf	$\text{CF}_3\text{CF}=\text{CH}_2$	2,3,3,3-tetrafluoroprop-1-ene	754-12-1	2057041
R1234zc	$\text{CHF}_2\text{CH}=\text{CF}_2$	1,1,3,3-tetrafluoroprop-1-ene	4556-24-5	10323104
R1234ze(E)	$\text{CF}_3\text{CH}=\text{CHF}$	(1E)-1,3,3,3-tetrafluoroprop-1-ene	29118-24-9	4647426
R1234ze(Z)	$\text{CF}_3\text{CH}=\text{CHF}$	(1Z)-1,3,3,3-tetrafluoroprop-1-ene	29118-25-0	9291157
R1243yc	$\text{CH}_3\text{CF}=\text{CF}_2$	1,1,2-trifluoroprop-1-ene	563-85-9	13460334
R1243ye(E)	$\text{CH}_2\text{FCF}=\text{CHF}$	(1E)-1,2,3-trifluoroprop-1-ene	1237522-12-1	16023665
R1243yf	$\text{CHF}_2\text{CF}=\text{CH}_2$	2,3,3-trifluoroprop-1-ene	158664-13-2	14074491
R1243zc	$\text{CH}_2\text{FCH}=\text{CF}_2$	1,1,3-trifluoroprop-1-ene	58777-31-4	19054626
R1243ze(E)	$\text{CHF}_2\text{CH}=\text{CHF}$	(1E)-1,3,3-trifluoroprop-1-ene	100922-85-8	4896157
R1243zf	$\text{CF}_3\text{CH}=\text{CH}_2$	3,3,3-trifluoroprop-1-ene	677-21-4	12151

Table 2. Normal boiling point temperatures (K), and source of information, for refrigerants listed in Table 1.

Refrigerant	Primary Publicly Available Source	Chemical Supplier ¹⁰	NIST Webbook ¹¹	Other Secondary Publicly Available Sources
R1225yc	275.15 ¹	274.15 to 275.15	—	—
R1225ye(E)	258.15 ²	255.15	—	—
R1225ye(Z)	253.15 ³	255.15	—	—
R1225zc	251.35 ⁴	252.15	252	—
R1234yc	—	—	—	—
R1234ye(E)	251.15 ⁵	—	—	—
R1234ye(Z)	—	—	—	—
R1234yf	243.71 ⁶	244.85	—	245.15 ⁸ ; 243.7 ¹² ; 244.15 ¹³ ; 243.75 ¹⁴
R1234zc	—	—	—	—
R1234ze(E)	254.21 ⁷	257.15	—	254.15 ⁸ ; 254.2 ¹² ; 254.15 ¹⁵
R1234ze(Z)	282.15 ⁸	284.15 to 285.15	—	—
R1243yc	—	—	—	—
R1243ye(E)	—	—	—	—
R1243yf	—	—	—	—
R1243zc	—	—	—	—
R1243ze(E)	—	—	—	—
R1243zf	247.95 ⁹	255.15 to 257.15	255	247.15 ¹⁶

¹Hulse *et al.* (2012)²Knapp *et al.* (2008)³Miller *et al.* (2008)⁴Miller *et al.* (2007)⁵Luly and Singh (2008)⁶Fedele *et al.* (2011)⁷Di Nicola *et al.* (2012)⁸Mukhopadhyay *et al.* (2008)⁹Low (2009)¹⁰www.snyquestlabs.com¹¹webbook.nist.gov¹²Lemmon *et al.* (2010)¹³Spatz and Minor (2008)¹⁴Honeywell (2011)¹⁵Honeywell (2008)¹⁶Pham *et al.* (2010)

Table 3 lists the known NBP and estimated NBP for the isomers of Table 1. In this table, unlike in Tables 1 and 2, the isomers are grouped by the “accuracy” of the NBP. The “well-established” column lists values from public domain sources for nine of the isomers. The next column displays estimated NBP values for the 17 isomers by the

authors of this paper based on the modified Joback method (REF) and the following two columns display estimated NBP for the 17 isomers based on estimation techniques implemented in ChemSpider (www.chemspider.com).

The mean absolute error values of the estimated NBP are 3.8 % for the modified Joback method, 4.1 % for the ACD/PhysChem method, and 3.1 % for the EPS Suite method. When the averages of the three methods (the last column) are compared to the known NBP, the resulting mean absolute error value of 2.7 % is lower than any of the three methods individually. Thus, for the third grouping of isomers (the eight without well-established NBP), the average estimated values (the last column) are used in the development of the estimates of critical state properties, ideal gas specific heats at constant pressure, and acentric factors, which are used in the development of the EoS. Whereas, for the first grouping of isomers (R1234yf and R1234ze(E)), the critical state properties, ideal gas specific heats at constant pressure, and acentric factors are from well-established sources, and for the second groupings of isomers, the well-established NBP are used in the development of the estimates of critical state properties, ideal gas specific heats at constant pressure, and acentric factors.

Table 3. Normal boiling point temperatures (K), including estimated values, of the refrigerants of Table 1, where they have been grouped by how much is thermodynamically “known” about them.

Refrigerant	Well-established	Modified Joback (estimation technique)	ACD/PhysChem (estimation technique)	EPS Suite (estimation technique)	Average of three estimating techniques
R1234yf	243.71	251.59	236.85	238.43	242.29
R1234ze(E)	254.21	259.19	253.86	257.02	256.69
R1225yc	275.15	274.57	267.07	255.58	265.74
R1225ye(E)	258.15	264.68	258.46	252.10	258.41
R1225ye(Z)	253.15	264.68	258.46	252.10	258.41
R1225zc	251.35	264.68	236.76	252.10	251.18
R1234ye(E)	251.15	269.08	289.77	260.48	273.11
R1234ze(Z)	282.15	259.19	–	257.02	258.11
R1243zf	247.95	246.10	256.15	243.44	248.56
R1234yc	–	269.40	281.97	269.65	273.67
R1234ye(Z)	–	269.08	289.77	260.48	273.11
R1234zc	–	269.08	259.05	260.48	262.87
R1243yc	–	263.79	264.01	266.94	264.91
R1243ye(E)	–	263.91	304.93	274.48	281.11
R1243yf	–	255.99	262.70	246.95	255.21
R1243zc	–	263.91	265.86	274.48	268.08
R1243ze(E)	–	263.59	277.29	265.36	268.75

Table 4 lists the critical state properties, ideal gas specific heats at constant pressure and acentric factors for the refrigerants of Table 3, which are needed to construct EoS, such as P-R and ECS, to name two. For R1234yf and R1234ze(E), the values are taken from Lemmon *et al.* (2010) and for the others they are estimates based on the methodology described by Brown *et al.* (2010). In fact, the values for R1225ye(E), R1225ye(Z), R1225zc, R1234ye(E), R1234ze(Z), and R1243zf are taken from Brown *et al.* (2010) and for the other nine isomers the values are estimated for the first time in this paper using the average estimated NBP (the last column of Table 3).

Although details are not provided or described herein, P-R EoS were constructed for the 17 isomers from the data of Table 4. These EoS were then used to generate vapor pressure estimates from 250 K to the critical temperature for each of the 17 isomers. These estimates were then fitted to a Wagner-type vapor pressure correlation described in Equation (1). Finally, the vapor pressures for the 17 isomers are plotted in Figure 1.

$$T_r \ln(P_r) = A_1 \tau + A_2 \tau^{1.5} + A_3 \tau^{2.5} + A_4 \tau^5 \quad (1)$$

with the constants given in Table 5 and where the reduced temperature $T_r = T/T_c$, the reduced pressure $P_r = P/P_c$, and $\tau = 1 - T_r$.

Table 4. Critical state properties, ideal gas specific heats at constant pressure, and acentric factors of the refrigerants of Table 3.

Refrigerant	T_c (K)	P_c (kPa)	ρ_c (kg/m ³)	c_p° @ $T_r = 0.8$ (kJ/kg ¹ ·K)	ω
R1234yf	367.9	3382	476	0.882	0.276
R1234ze(E)	382.5	3636	489	0.885	0.313
R1225yc	412.3	3625	517	0.820	0.336
R1225ye(E)	386.8	3401	517	0.789	0.313
R1225ye(Z)	379.3	3335	517	0.778	0.305
R1225zc	376.7	3312	517	0.774	0.303
R1234ye(E)	379.9	3534	473	0.828	0.290
R1234ze(Z)	426.8	3970	473	0.906	0.333
R1243zf	378.8	3740	423	0.934	0.274
R1234yc	413.9	3851	473	0.878	0.322
R1234ye(Z)	413.1	3843	473	0.876	0.321
R1234zc	397.6	3699	473	0.854	0.307
R1243yc	404.6	3996	423	0.940	0.297
R1243ye(E)	429.4	4240	423	0.979	0.318
R1243yf	389.8	3850	423	0.952	0.284
R1243zc	409.5	4044	423	0.947	0.302
R1243ze(E)	410.5	4054	423	0.947	0.303

Table 5. Constants for equation (1) based on vapor pressure estimates generated from P-R EoS for refrigerants other than R1234yf and R1234ze(E). The constants for R1234yf and R1234ze(E) are taken from Fedele *et al.* (2011) and Di Nicola *et al.* (2012), respectively.

Refrigerant	A_1	A_2	A_3	A_4
R1234yf	-7.3442	1.4957	-1.7726	-4.9249
R1234ze(E)	-7.5046	1.5524	-2.2353	-4.1018
R1225yc	-7.3370	0.6516	-0.9704	-5.4550
R1225ye(E)	-7.2181	0.5843	-0.6949	-6.0777
R1225ye(Z)	-7.1758	0.5600	-0.5994	-6.3271
R1225zc	-7.1643	0.5499	-0.5649	-6.4567
R1234ye(E)	-7.0904	0.5087	-0.4542	-6.3870
R1234ze(Z)	-7.3349	0.6992	-1.0632	-5.0130
R1243zf	-7.0289	0.5451	-0.4524	-6.0144
R1234yc	-7.2672	0.6375	-0.8915	-5.3120
R1234ye(Z)	-7.2714	0.6588	-0.9174	-5.2534
R1234zc	-7.1969	0.6106	-0.7444	-5.6036
R1243yc	-7.1550	0.6212	-0.7382	-5.3056
R1243ye(E)	-7.2697	0.6989	-1.0038	-4.8359
R1243yf	-7.0844	0.5786	-0.5792	-5.6664
R1243zc	-7.1802	0.6429	-0.8015	-5.1756
R1243ze(E)	-7.1831	0.6410	-0.8054	-5.1697

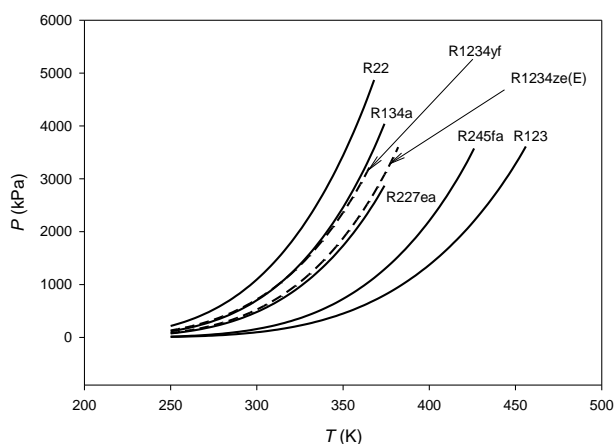


Figure 1a. Vapor pressures of fluorinated propene isomers with well-established thermodynamic properties. Included in the figure are some common HCFC and HFC refrigerants.

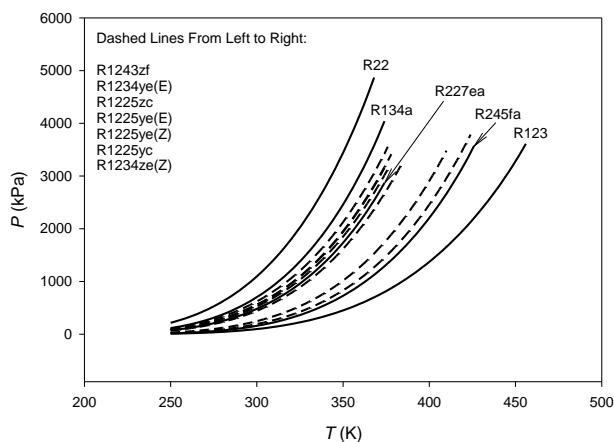


Figure 1b. Vapor pressures of fluorinated propene isomers based on well-established normal boiling point temperatures. Included in the figure are some common HCFC and HFC refrigerants.

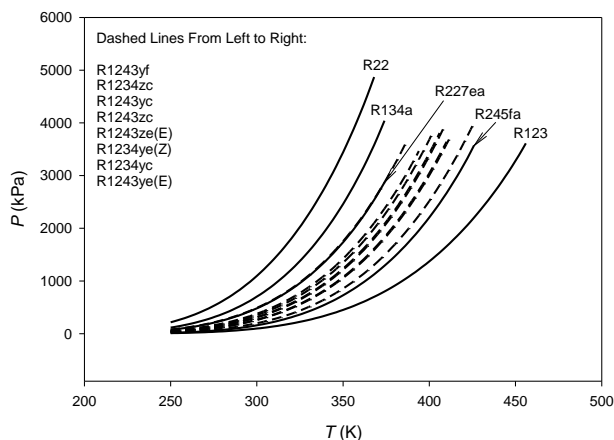


Figure 1c. Vapor pressures of fluorinated propene isomers based on estimated normal boiling point temperatures. Included in the figure are some common HCFC and HFC refrigerants.

5. CONCLUSIONS

This paper critically reviewed vapor pressure data and vapor pressure models for seventeen hydrofluoroolefins, including R1234yf and R1234ze(E), and presented Wagner-type vapor pressure correlations for the seventeen hydrofluoroolefins. At 273 K, the highest pressure refrigerant among the seventeen is R1234yf and the lowest pressure refrigerant among the seventeen is R1234ze(Z). At 273 K, the vapor pressure of R1234ze(E) is greater than eleven of the other sixteen isomers. At 273 K, the vapor pressure of R1234yf is 45.9 % greater than the vapor pressure of R1234ze(E), 364 % greater than the vapor pressure of R1234ze(Z), and 7.9 % greater than the vapor pressure of R134a.

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